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TECHNICAL REPORT MIT-50-PU

VISCOSITY OF NITROGEN, HELIUM,
HYDROGEN, AND ARGON FROM -100° TO 25°C
UP TO 150-250 ATMOSPHERES

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TABLE OF CONTENTS

<u>Section</u>	<u>Page</u>
Abstract.	1
Introduction.	2
Apparatus and Procedures.	3
Experimental Results.	5
Density Dependence of Viscosities	7
Accuracy.	11
Acknowledgments	12
References.	13
Figure Captions	16

Viscosity of Nitrogen, Helium, Hydrogen, and Argon
from -100° to 25°C up to 150-250 Atmospheres*

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The range of a previously developed capillary-flow viscometer has been extended to -100°C , and the techniques of obtaining and determining the attainment of steady state have been improved. The viscosities of nitrogen, helium, hydrogen, and argon have been measured by an absolute method from -100° or -90° to 25°C and up to 150-175 atm (250 atm for N_2 at 25°C). The accuracy is estimated to be 0.1 - 0.2%, but is somewhat worse for Ar at -100°C . A number of empirical and theoretical analyses of the viscosity-density dependence have been made; while inconclusive, the evidence tends not to support the presence of a logarithmic term in the density expansion. Correlations of the second and the third virial coefficients of viscosity have been made.

We report here a series of absolute measurements of the viscosity of four gases as a function of temperature and density. The purpose of this work is to obtain experimental data on this transport property in regions where such information is unavailable or insufficient; to investigate the temperature dependence of the virial coefficients for the viscosity; and to provide, if possible, a test of proposed theories on the density expansion.

We have in previous publications^{1,2} described our viscometer and given results for a number of gases at temperatures from -50° to 150°C . The present work extends the temperature range to -90°C for N_2 and He and to -100°C for H_2 and Ar, and repeats the previous measurements at -50° and 25°C for more closely spaced values of the density. As before, the upper pressure limits are in the range 150-175 atm, except for N_2 at 25°C , which we studied to about 250 atm. The precision of the data is better than 0.1%, and the results should be accurate to 0.1 - 0.2%, with the exception of Ar at -100°C (see discussion below).

At the time of publication of Ref. 1, there were hardly any other published measurements of the viscosity of gases below 0°C and above 1 atm, and none of comparable accuracy. This situation has improved in recent years, especially with Kao and Kobayashi's³ work on N_2 and He from -90° to 50°C and Diller's⁴ on p- H_2 below 100°K , as well as some extensive measurements of much lesser accuracy mentioned below. However, none of these papers contains enough low-density data to give good values of the second virial coefficient of viscosity which is of theoretical interest.⁵⁻⁹ For this reason the present measurements have been extended to lower pressures than our previous work.^{1,2}

APPARATUS AND PROCEDURES

The apparatus and technique of measurement have been described in Ref. 1, and only some changes need be noted here; for a more extensive discussion, see Ref. 10. The apparatus is a constant-volume capillary-flow viscometer, in which an injector at 25°C drives gas at a known flow rate through a capillary at the experimental temperature, the resulting pressure difference being determined with a differential manometer.

The only major change is the addition of a fused-quartz precision pressure gage (Texas Instruments Inc., Houston, Texas) in parallel with the differential manometer. This instrument registers a pressure difference by deflection of a quartz Bourdon tube. We had hoped to use it for absolute measurements, but found that the variation of the index of refraction of a gas with pressure affects the optical path in the instrument, so that its calibration varies with pressure. We therefore continued to use the mercury manometer, but made use of the pressure gage to monitor the approach to steady state.

Steady state is defined here as invariance with time of the pressure difference across the capillary during a run. Strictly speaking, such steady-state behavior is impossible in a constant-volume apparatus, since the pressure difference steadily increases with time;¹¹ however, we have shown¹⁰ that this increase is negligible for the dimensions of our apparatus. A more serious problem is the relaxation of the transient process set in motion when a run begins: in particular, the rapid imposition of the pressure difference is an essentially adiabatic process, producing a temperature difference across the capillary. For most of the measurements reported here, we in fact observed the attainment of steady state (constant within c. 0.001

in. Hg) with the pressure gage as noted above. In addition, a rough but adequate model of the transient behavior has been derived¹⁰ and approximately confirmed experimentally, to the extent that we believe steady state to have been reached in virtually all the previous measurements with this apparatus. (This applies even for injector flow rates up to 0.05 cm³/sec, although we now work only at ≤ 0.03 cm³/sec.)

Three capillaries were used in the present work, all about 36 cm long and 0.019 cm in diameter. The last two were calibrated by the electrical-resistance method, which we have found to be both more convenient and more precise than the gravimetric method.¹² More careful analysis of our previous techniques with the latter method led us to the conclusion¹⁰ that the viscosity values in Refs. 1 and 2 should be decreased by about 0.12%.¹³

For each capillary the value of the kinetic-energy constant m was determined in the same way as before¹, except that individual runs were weighted in accordance with their a priori precision.¹⁰ We again took an upper Reynolds-number limit of 1600, below which we found no correlation of m with Reynolds number or other variables. For the three capillaries the values of m obtained were 1.199 ± 0.013 , 1.162 ± 0.016 , and 1.199 ± 0.025 ; these values were used to calculate the viscosities in Table I.

EXPERIMENTAL RESULTS

Our measured viscosities are given in Table I and plotted in Figs. 1-4; the deviations listed in the table indicate primarily the scatter of the measurements at each pressure. The paragraphs which follow give our density sources and some discussion of the results. More extensive comparisons with previous work can be found in Ref. 10.

Nitrogen

Nitrogen¹⁴ was studied up to 253 atm at 25°C; at -50° and -90°C, as for the other gases, limits were chosen in the range 150-175 atm. The results are shown in Fig. 1. The densities used were those of Michels *et al.*¹⁵ at 25°C and of Canfield *et al.*¹⁶ at lower temperatures. Our results are in good agreement¹⁷ with our previous work¹, and thus are still about 0.4% lower than those of Kestin and Leidenfrost¹⁸ and Michels and Gibson¹⁹ at 25°C and moderate pressures. Agreement with Kao and Kobayashi³ at -50° and -90°C is within their precision. On the other hand, our disagreement with Ross and Brown²⁰ is as reported before,¹ and Filippova and Ishkin's²¹ measurements deviate from ours in the same way; Goldman's²² data at -61° and -78.5°C are 1-6% higher than values interpolated from ours; and extrapolation of our -90°C curve indicates that Zhdanova's²³ results for $\rho \geq 0.38$ g/cm³ are as much as 20% higher.

Helium

Helium¹⁴ was also studied at 25°, -50°, and -90°C; the results are shown in Fig. 2. The densities are based on the work of Miller *et al.*²⁵ at 25°C and of Canfield *et al.*¹⁶ elsewhere. We are again in good agreement with

our previous work¹ and about 0.3% lower than the results of Kestin et al.¹⁸ around 25°C. At -50° and -90°C, Kao and Kobayashi's³ results average 0.2-0.3% lower than ours. Golubev and Gnezdilov²⁶ obtain viscosities consistently steeper in the density than all the above work, but closely resembling those of Ross and Brown²⁰, which we regard as unreliable.¹ Kestin's results, Kao and Kobayashi's, and our's all tend to confirm the existence of very shallow minima in the viscosity-density curves.

Hydrogen

Hydrogen²⁷ was studied at 25°, -50°, and -100°C; the results are shown in Fig. 3. The densities were all taken from Michels et al.²⁸ Agreement with our previous work² was excellent at 25°C and fairly good (the present data averaging slightly higher) at -50°C. Near 25°C, Kestin and Leidenfrost's¹⁸ results are about 1% higher than ours, but the recent work of Kestin and Yata²⁹ is only about 0.5% higher, being thus in good agreement with Michels, Schipper, and Rintoul.³⁰ There is no other high-precision work in our lower-temperature range; both Golubev and Shepeleva³¹ and Rudenko and Slyusar³² obtain results which increase with density somewhat more rapidly than ours or Michels'.

Argon

Argon³³ was also studied at 25°, -50°, and -100°C; the results are shown in Fig. 4. The densities were obtained from Michels, Wijkers and Wijkers³⁴ at 25°C, and from Michels, Levelt, and de Graaff³⁵ at the lower temperatures. Agreement with our earlier work¹ is again fairly good. As before, at 25°C and moderate densities we are about 0.5% below Kestin et al.¹⁸ and 0.2% above Michels, Botzen, and Schuurman.³⁶ Filippova and Ishkin's²¹ low-temperature data deviate from ours in the same way as for H₂. Our -100°C isotherm requires

special discussion, which must be postponed until the questions of density dependence have been considered.

DENSITY DEPENDENCE OF VISCOSITIES

For each isotherm the viscosity was fitted to a least-squares power series in the density:

$$\eta = a + bp + cp^2 + \dots \quad (1)$$

These fits were tried for polynomials of different degrees, the "best" fit (to all the points of the isotherm) being chosen by a variety of statistical tests;³⁷ the constants of the resulting series are listed in Table II. The standard deviations (σ) of the fits are in each case well under 0.2%, except for N_2 at $-90^\circ C$ and Ar at $-100^\circ C$ (for both of which the individual viscosities also have quite high deviations), thus confirming our estimate of precision (see below).

The "best" fits just described, however, do not necessarily yield the best values of the virial coefficients, as has been shown by Hanley, McCarty, and Senners.³⁸ These authors found, by fitting series to data subsets of increasing density range, that (for example) a good linear fit could be obtained up to some limiting density ρ_L , beyond which the data deviated from linearity, but that good coefficients for a quadratic fit were not obtained below some density appreciably greater than ρ_L . Most of our isotherm ranges fall in such intermediate regions. We have therefore carried out such subset fits for our data, and the resulting estimates for the best of values of a , b , c are given in Table III for N_2 and Ar; the

scatter was too great for satisfactory results by this technique for He and H₂. The values in Table III should thus be taken as our best estimates of these virial coefficients, while those in Table II are best for interpolation over our entire density range.

For N₂ and Ar fits were also made to the series

$$\eta = a + bp + \rho^2 (c' + d' \ln p), \quad (2)$$

which has been proposed on theoretical grounds.³⁸ In each case¹⁰ the goodness of fit was no better or (usually) slightly worse than for the power series (1) with an equal number of coefficients. Our work thus offers no support for the logarithmic-term hypothesis, whereas Hanley, McCarty, and Sengers⁹ did find some support in an analysis based primarily on thermal conductivity data.

In addition to our individual-isotherm fits, further tests were made of the hypothesis¹ that the residual viscosity is a function of density only, i.e., that

$$\eta(T, \rho) = \eta(T, 0) + \Delta\eta(\rho) \quad (3)$$

This is an empirical correlation, the breakdown of which at low temperatures and high densities has been fairly well established.^{4,23} However, where it is valid it not only is quite useful for purposes of estimation, but holds to a remarkable degree of accuracy. Such fits were therefore made, as described in Ref. 1, for each of the gases studied; the standard deviations of the "best" fits are 0.26 mP for N₂, 0.14 for He, 0.14 for H₂, 1.41 for Ar (0.46 with -100°C omitted). (See Ref. 10 for the constants in these series and detailed comparison graphs showing to what extent the isotherms are in fact parallel.) Except for Ar, these results are quite consistent with the validity of Eq. (3) for our gases, within our experimental accuracy.

The Ar results, however, are problematical. Even the 25° and -50°C results are definitely not parallel within their own precision (the 25°C data being parallel to Kestin's and Michels' results), but the discrepancy here is only of the order of 1 μ P. The -100°C data, in contrast, dip as much as 4% below the values predicted from Eq. (3) and Michels' $\Delta\eta(\rho)$, then rise more steeply at higher densities. (It may be significant that the greatest discrepancy, nearly 10 μ P, is at 0.5 g/cm³, quite close to the critical density of 0.531 g/cm³.) The -50°C deviations, while much smaller, are in the same direction (as are the previous rough results¹ at -78.5°C). As noted above, Eq. (3) is known to fail in the low-temperature liquid-density range, but the previous evidence^{4,23} indicates that the departure is in the direction of higher values of $\Delta\eta(\rho)$, the opposite of what we observe here; no effect of either kind appears in the present N₂ data, at similar reduced temperatures and densities. The poor precision of the -100°C data is far too small an effect to account for this discrepancy, and further low-temperature Ar measurements are clearly desirable.

To return to the individual-isotherm fits, we have reduced the values of the second virial coefficient of viscosity, b , from Table III for N₂ and Ar, Table II for H₂ and He for comparison with the theories of Kim and Ross⁸ and Hoffman and Curtiss.⁷ The reduced values, b_{η}^* , are shown in Fig. 5, along with the two theoretical curves; see Refs. 8 for comparison with several other theoretical models,^{5,6,39} none of which fit the experimental data well. Also shown in Fig. 5 are the values of b_{η}^* from the data of Kestin and Leidenfrost,¹⁸ to which the parameters in Kim and Ross's theory were fitted; the present values are in general higher, but the shape of the curve remains unchanged.⁴⁰ The theory of Kim and Ross seems reasonably

correct, except for its failure to predict the negative values of b for He; on the other hand, that of Hoffman and Curtiss agrees well with the He data but fails at lower reduced temperatures. These results can be explained as due to Kim and Ross taking inadequate account of repulsive interactions and Hoffman and Curtiss's neglect of orbiting, effects important at high and low reduced temperatures respectively. The plateau predicted by Kim and Ross for $(1/T^*) \geq 0.3$ presumably is related to the nearly temperature independent residual viscosity discussed above, although the latter appears to apply even at densities where higher virial coefficients are dominant.

In connection with the point just made, a correlation of the third virial coefficient should be useful; to our knowledge, this has not been attempted before. While no theory for the value of c exists, the appropriate reduction is easily made by dimensional analysis: since we have⁸

$$b_{\eta}^* (T^*) = \frac{(6/\sigma)}{(\epsilon/m)^{\frac{1}{2}}} b_{\eta} \quad , \quad (4)$$

where σ and ϵ are Lennard-Jones parameters and $T^* = kT/\epsilon$, it is clear that a dimensionless c_{η}^* should be given by

$$c_{\eta}^* (T^*) = \frac{(1/\sigma^4)}{(\epsilon/m^3)^{\frac{1}{2}}} c_{\eta} \quad . \quad (5)$$

If σ is given in Å and (ϵ/k) in $^{\circ}\text{K}$, the reduction factor $\sigma^4(\epsilon/m^3)^{\frac{1}{2}}$ equals $54.92 \sigma^4 (\epsilon/k M^3)^{\frac{1}{2}} \mu\text{P-cm}^6/\text{g}^2$, where M is the molecular weight. Values of c_{η}^* were therefore obtained from our data as just described for b_{η}^* , the results being plotted in Fig. 6. Once again a plateau is obtained over most of the temperature range.

ACCURACY

Our previous estimate¹ of experimental precision was 0.001 in. Hg in the pressure difference. Although in our low-temperature work we were frequently restricted (by steady-state considerations) to pressure differences much less than an inch, the reproducibility of the viscosities (Table I) remained better than 0.1% in nearly all cases (even for H₂ at -100°C, where none of the pressure differences exceeded 0.4 in. Hg). The exceptions to the above statement are of course N₂ at -90°C and Ar at -100°C. As mentioned in the previous section, the standard deviations of our isotherm least-squares fits are consistent with these estimates of precision. Our average precision is thus about 0.05% near room temperature, but becomes worse than 0.1% at low temperatures and high densities (perhaps due to lack of thermal equilibrium in the apparatus).

Our accuracy should be somewhat worse than this. For our low-temperature N₂ and He measurements, there is the possibility¹⁰ of errors larger than 0.1% in the density ratios and thus the viscosities, due to our having combined densities from two different sources; the single-source H₂ and Ar density ratios should be good to about 0.02%. Combining this with other known sources of error in the calibrations and measurements, we estimate our accuracy as 0.1 - 0.2%.

However, accuracy can be established objectively only by comparison with other work, and the situation in this respect is unsettled. Our agreement with previous work with this apparatus^{1,2} is within the limits just stated, but at -50°C there are some apparently systematic discrepancies¹⁰ at the outer range of these limits. The most precise other work available

for comparison is that of Kestin et al.,^{18,29} done with oscillating-disc viscometers, and the discussions above have indicated that our results are consistently several tenths of a percent lower than Kestin's; the possibility of flaws in the theory of either or both instruments may be worth investigating. We are in good agreement with Kao and Kobayashi³ for N₂ and slightly higher for He, while our discrepancies with Michels et al.^{19,30,36} are not consistent (both of these being capillary measurements). There are no other dense-gas data sufficiently accurate for useful comparison, but it should be noted that our 1-atm viscosities (based on Table II), except for He, are consistently an average of about 0.5% lower than the bulk of previous work¹⁰; since we deliberately extended our work to lower densities than before, this can no longer be attributed to extrapolation error. The weight of the evidence suggests the possibility of systematic errors, but is not sufficiently consistent to be conclusive.

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References

- * Supported in part by Project Squid, Office of Naval Research, and the National Science Foundation.
- † Present address: Pratt and Whitney Aircraft, Div. United Aircraft Corp., East Hartford, Connecticut.
1. G. P. Flynn, R. V. Hanks, N. A. Lemaire, and J. Ross, J. Chem. Phys. 38, 154 (1963). For corrections to the data given in Refs. 1-2, see comments below, especially Ref. 13, or Ref. 10 for more details.
 2. A. K. Barua, M. Afzal, G. P. Flynn, and J. Ross, J. Chem. Phys. 41, 374 (1964). In Table V of this paper, b for H₂ at 75°C should read 74.6 rather than 7.5.
 3. J. T. F. Kao and R. Kobayashi, J. Chem. Phys. 47, 2836 (1967).
 4. D. E. Diller, J. Chem. Phys. 42, 2089 (1965).
 5. D. E. Stogryn and J. O. Hirschfelder, J. Chem. Phys. 31, 1545 (1959); 33, 942 (1960).
 6. R. F. Snider and C. F. Curtiss, Phys. Fluids 1, 122 (1958); C. F. Curtiss, M. B. McElroy, and D. K. Hoffman, Int. J. Eng. Sci. 3, 269 (1965).
 7. D. K. Hoffman and C. F. Curtiss, Phys. Fluids 8, 890 (1965).
 8. S. K. Kim and J. Ross, J. Chem. Phys. 42, 263 (1965); S. K. Kim, G. P. Flynn, and J. Ross, ibid. 43, 4166 (1965).
 9. H. J. M. Hanley, R. D. McCarty, and J. V. Sengers, J. Chem. Phys. 50, 857 (1969).
 10. J. A. Gracki, Ph.D. thesis, Brown University, 1969.
 11. K. R. Van Doren, R. A. Guereca, H. P. Richardson, and D. Cummins, (2 articles submitted to J. Appl. Mech.)

12. For these methods, see J. F. Swindells, J. R. Coe, Jr., and T. B. Godfrey, J. Res. Nat. Bur. Stand. (U.S.) 48, 1 (1952).
13. In addition, the measurements reported at -50°C in Refs. 1 and 2 were actually made at -49.90°C , so that these values should be corrected by an additional amount. The correction is in most cases only slightly pressure-dependent, and averages $-0.05\text{ }\mu\text{P}$ for H_2 , -0.07 for D_2 , -0.11 for He , -0.12 for N_2 and CO , -0.17 for Ar , and -0.19 for Ne ; for CH_4 (near its critical temperature) the pressure variation is too great for accurate estimation of the correction.
14. Purity 99.998%, from Air Reduction Co.
15. A. Michels, R. J. Lunbeck, and G. J. Wolkers, Physica 17, 801 (1951).
16. F. B. Canfield, T. W. Leland, and R. Kobayashi, J. Chem. Eng. Data 10, 92 (1965). These densities (for both N_2 and He) differ appreciably from those used in Ref. 1. For reduction of our earlier data to the same basis as the present data, see Ref. 10; the values thus obtained are used in Figs. 1-2 and in the discussion of comparative results below.
17. "Good agreement" with our previous work, for all the gases in this paper, means agreement within the combined postulated limits of accuracy (see section "Accuracy" below), after correction^{13,16} of the earlier data.
18. J. Kestin and W. Leidenfrost, Physica 25, 1033 (1959); see also several more recent papers by Kestin et al., primarily devoted to mixtures at 20° - 30°C .
19. A. Michels and R. O. Gibson, Proc. Roy. Soc. (London) A134, 288 (1931).
20. J. F. Ross and G. M. Brown, Ind. Eng. Chem. 49, 2026 (1957).
21. G. P. Filippova and I. P. Ishkin, Inzh.-Fiz. Zh., Akad. Nauk Belorus. SSR 4, 105 (1961).
22. K. Goldman, Physica 29, 499 (1963).
23. N. F. Zhdanova, Zh. Eksp. Teor. Fiz. 31, 14 (1956); Sov. Phys.-JETP 4, 19 (1957).

24. Purity 99.995%, from Matheson Co.
25. J. E. Miller, L. W. Brandt, and L. Stroud, U. S. Bur. Mines, Rep.-Invest., No. 5845 (1961).
26. J. F. Golubev and N. E. Gnezdilov, *Gazov. Prom.* 10, (12), 38 (1965).
27. Purity 99.95%, from Matheson Co.
28. A. Michels, W. de Graaff, J. Wassenaar, J. M. H. Levelt, and P. Louwerse, *Physica* 25, 25 (1959).
29. J. Kestin and J. Yata, *J. Chem. Phys.* 49, 4780 (1968).
30. A. Michels, A. C. J. Schipper, and W. H. Rintoul, *Physica* 19, 1011 (1953).
31. J. F. Golubev and R. I. Shepeleva, *Gazov. Prom.* 11 (4), 54 (1966).
32. N. S. Rudenko and V. P. Slyusar, *Ukr. Fiz. Zh. (Russ. Ed.)* 13, 917 (1968).
33. Purity 99.998%, from Matheson Co.
34. A. Michels, Hub. Wijker, and Hk. Wijker, *Physica* 15, 627 (1949).
35. A. Michels, J. M. Levelt, and W. de Graaff, *Physica* 24, 659 (1958).
36. A. Michels, A. Botzen, and W. Schuurman, *Physica* 20, 1141 (1954).
37. A. Michels, J. C. Abels, C. A. Ten Seldam, and W. de Graaff, *Physica* 26, 381 (1960); K. R. Hall and F. B. Canfield, *ibid.* 33, 481 (1967).
See Ref. 10 for details of the application to the present data.
38. A recent review of the theory on this subject is given by J. V. Sengers, *Boulder Lectures in Theoretical Physics*, vol. 9C, *Kinetic Theory* (Gordon and Breach, New York, 1967), 335.
39. D. Enskog, *Kgl. Sv. Vetenskapsakad. Handl.* [2] 63, No. 4 (1922).
40. A graph similar to Fig. 5, but including data from many other sources, is given by Hanley, McCarty and Sengers;⁹ although their analysis of the data was carried out largely by fitting to Eq. (2), the results appear qualitatively the same.

FIGURE CAPTIONS

- Fig. 1 Viscosity of nitrogen versus density: ● present results; ○ Flynn, Hanks, Lemaire, and Ross¹ (points nearly coinciding with present data indicated by ⊙); ✕ Kao and Kobayashi.³
- Fig. 2 Viscosity of helium versus density: ● present results; ○ Flynn, Hanks, Lemaire, and Ross¹; ✕ Kao and Kobayashi.³
- Fig. 3 Viscosity of hydrogen versus density: ● present results; ○ Barua, Afzal, Flynn, and Ross²; △ Diller.⁴
- Fig. 4 Viscosity of argon versus density: ● present results; ○ Flynn, Hanks, Lemaire, and Ross¹ (points nearly coinciding with present data indicated by ⊙).
- Fig. 5 Comparison of theoretical^{5,8} and experimental values of b_{η}^* , the reduced second virial coefficient of viscosity: present data, ● N₂, ■ H₂, ▼ H₂, ▲ Ar; Kestin and Leidenfrost,¹⁸ ○ N₂, □ He, ▽ H₂, △ Ar, ⊕ O₂, ⊖ D₂, + Ne, ✕ Kr, * Xe. See Refs. 8 for the method of calculating b_{η}^* ; the experimental values for the present data are from Table II for N₂ and Ar, Table III for He and H₂.
- Fig. 6 Values of c_{η}^* , the reduced third virial coefficient of viscosity, from our data: ● N₂, ■ He, ▼ H₂, ▲ Ar (from Table II for N₂ and Ar, Table III for He and H₂).

TABLE I. Measured Viscosities.

Pressure (atm)	Density (g/cm ³)	Viscosity (μ P)	Pressure (atm)	Density (g/cm ³)	Viscosity (μ P)
N₂, 25°C					
6.77	0.00776	177.93±0.04	5.28	0.01000	119.32±0.11
20.06	0.02396	180.27±0.03	10.39	0.02000	119.65±0.06
"	"	180.14±0.02	20.16	0.04000	123.02±0.07
"	"	180.05±0.07	29.37	0.06000	127.60±0.34
41.68	0.04792	183.82±0.04	42.30	0.09000	133.46±0.13
52.20	0.06000	186.05±0.07	54.38	0.12000	140.76±0.18
62.52	0.07188	188.78±0.05	65.85	0.15000	149.55±0.25
83.71	0.09585	193.97±0.06	76.95	0.18000	159.19±0.08
105.24	0.11981	200.19±0.07	87.94	0.21000	169.73±0.10
"	0.11981	200.07±0.06	99.10	0.24000	182.50±0.20
123.81	0.14000	205.59±0.10	114.80	0.28000	201.06±0.22
127.33	0.14377	206.79±0.06	132.26	0.32000	222.42±0.13
142.71	0.16000	211.72±0.11	152.63	0.36000	247.19±0.17
152.23	0.18000	218.41±0.12	He, 25°C		
173.92	0.19169	222.62±0.08	6.13	0.001000	198.07±0.03
198.96	0.21565	231.72±0.09	12.30	0.002000	197.91±0.03
253.52	0.26358	251.66±0.08	24.75	0.004000	197.83±0.02
N₂, -50°C					
6.48	0.01000	141.75±0.03	37.35	0.006000	197.76±0.07
12.85	0.02000	142.91±0.11	56.53	0.009000	197.84±0.03
25.29	0.04000	145.96±0.14	76.07	0.012000	197.88±0.03
37.40	0.06000	149.52±0.08	95.98	0.015000	197.92±0.03
49.24	0.08000	153.68±0.11	116.25	0.018000	197.95±0.03
60.89	0.10000	158.23±0.12	136.90	0.021000	198.12±0.03
72.43	0.12000	163.31±0.13	157.93	0.024000	198.27±0.07
83.92	0.14000	168.89±0.08			
95.46	0.16000	175.13±0.21			
"	"	175.03±0.17			
107.13	0.18000	181.77±0.15			
119.03	0.20000	188.96±0.19			
131.26	0.22000	196.89±0.21			
143.93	0.24000	204.92±0.15			
157.16	0.26000	214.11±0.13			
171.10	0.28000	223.62±0.15			

TABLE I. (continued)

Pressure (atm)	Density (g/cm ³)	Viscosity (μ P)	Pressure (atm)	Density (g/cm ³)	Viscosity (μ P)
He, -50°C					
4.59	0.001000	162.95 \pm 0.05	50.01	0.004000	89.23 \pm 0.06
9.21	0.002000	162.73 \pm 0.03	63.00	0.005000	89.45 \pm 0.04
18.53	0.004000	162.66 \pm 0.12	76.19	0.006000	89.72 \pm 0.01
27.97	0.006000	162.58 \pm 0.14	89.59	0.007000	89.97 \pm 0.04
42.35	0.009000	162.64 \pm 0.03	103.22	0.008000	90.24 \pm 0.01
57.00	0.012000	162.59 \pm 0.02	117.08	0.009000	90.50 \pm 0.04
71.93	0.015000	162.74 \pm 0.05	138.31	0.010500	90.86 \pm 0.05
87.14	0.018000	162.94 \pm 0.06	160.10	0.012000	91.41 \pm 0.02
102.64	0.021000	163.00 \pm 0.07			
118.43	0.024000	163.16 \pm 0.02			
134.53	0.027000	163.25 \pm 0.03			
150.93	0.030000	163.38 \pm 0.07			
167.65	0.033000	163.61 \pm 0.15			
He, -90°C					
7.56	0.002000	142.78 \pm 0.03	4.56	0.000500	72.85 \pm 0.02
15.21	0.004000	142.55 \pm 0.09	9.14	0.001000	73.01 \pm 0.03
22.95	0.006000	142.51 \pm 0.04	18.40	0.002000	73.14 \pm 0.02
34.75	0.009000	142.41 \pm 0.07	27.77	0.003000	73.38 \pm 0.01
46.77	0.012000	142.41 \pm 0.07	37.27	0.004000	73.63 \pm 0.05
63.16	0.016000	142.69 \pm 0.03	46.90	0.005000	73.80 \pm 0.01
79.97	0.020000	143.05 \pm 0.05	56.67	0.006000	73.96 \pm 0.01
97.21	0.024000	143.27 \pm 0.12	66.58	0.007000	74.13 \pm 0.03
114.86	0.028000	143.68 \pm 0.08	76.64	0.008000	74.36 \pm 0.05
133.04	0.032000	144.06 \pm 0.12	86.65	0.009000	74.60 \pm 0.03
151.67	0.036000	144.86 \pm 0.14	102.48	0.010500	75.02 \pm 0.01
170.78	0.040000	145.29 \pm 0.05	118.50	0.012000	75.51 \pm 0.03
			134.92	0.013500	76.15 \pm 0.04
			151.78	0.015000	76.78 \pm 0.04
			169.10	0.016500	77.52 \pm 0.06
H₂, 25°C					
6.09	0.000500	88.67 \pm 0.04			
12.23	0.001000	88.72 \pm 0.03			
24.63	0.002000	88.81 \pm 0.03			
37.23	0.003000	89.04 \pm 0.03			

TABLE I. (continued)

Pressure (atm)	Density (ρ/cm^3)	Viscosity (μP)	Pressure (atm)	Density (ρ/cm^3)	Viscosity (μP)
H_2 , -100°C					
7.08	0.001000	01.27 \pm 0.02	4.54	0.01000	176.41 \pm 0.02
14.23	0.002000	01.47 \pm 0.02	9.00	0.02000	177.36 \pm 0.04
21.43	0.003000	01.68 \pm 0.02	17.68	0.04000	179.78 \pm 0.11
32.43	0.004500	01.96 \pm 0.03	26.06	0.06000	182.64 \pm 0.06
43.60	0.006000	02.51 \pm 0.02	34.15	0.08000	185.71 \pm 0.11
54.97	0.007500	02.95 \pm 0.06	41.98	0.10000	189.15 \pm 0.10
"	"	03.03 \pm 0.02	49.57	0.12000	192.85 \pm 0.07
65.57	0.009000	03.71 \pm 0.07	64.10	0.16000	200.92 \pm 0.09
"	"	03.56 \pm 0.03	77.90	0.20000	210.35 \pm 0.10
78.41	0.010500	04.06 \pm 0.01	91.14	0.24000	220.98 \pm 0.15
90.52	0.012000	04.55 \pm 0.04	103.96	0.28000	232.53 \pm 0.17
107.10	0.014000	05.26 \pm 0.04	116.60	0.32000	245.24 \pm 0.16
124.23	0.016000	05.98 \pm 0.06	129.19	0.36000	259.12 \pm 0.15
141.97	0.018000	06.98 \pm 0.07	141.92	0.40000	274.60 \pm 0.17
160.36	0.020000	08.02 \pm 0.02	155.62	0.44000	290.93 \pm 0.21
Ar , 25°C					
7.30	0.01198	226.61 \pm 0.03	168.72	0.48000	308.85 \pm 0.18
14.54	0.02396	228.28 \pm 0.09			
28.85	0.04792	231.38 \pm 0.08			
42.95	0.07189	235.44 \pm 0.05			
56.87	0.09585	239.66 \pm 0.14			
70.66	0.11981	244.22 \pm 0.09			
84.35	0.14377	249.26 \pm 0.10			
111.60	0.19170	260.72 \pm 0.10			
138.93	0.23962	273.40 \pm 0.11			
166.70	0.28754	287.24 \pm 0.10			

TABLE I. (continued)

Pressure (atm)	Density (g/cm ³)	Viscosity (μP)
Ar, - 100°C		
6.89	0.02000	139.82 ± 0.23
13.33	0.04000	141.84 ± 0.15
19.36	0.06000	144.51 ± 0.19
19.36	0.06000	144.84 ± 0.31
27.64	0.09000	148.39 ± 0.08
35.09	0.12000	153.72 ± 0.19
43.85	0.16000	161.32 ± 0.06
51.44	0.20000	169.26 ± 0.27
59.56	0.25000	181.76 ± 0.17
66.45	0.30000	195.60 ± 0.20
72.43	0.35000	210.98 ± 0.48
77.78	0.40000	229.72 ± 0.14
82.79	0.45000	251.15 ± 0.98
87.76	0.50000	273.23 ± 0.19
93.01	0.55000	299.55 ± 0.72
98.96	0.60000	329.45 ± 0.45
106.12	0.65000	360.02 ± 0.23
115.17	0.70000	396.55 ± 0.15
126.96	0.75000	436.04 ± 0.40
142.61	0.80000	479.25 ± 0.52
158.86	0.84000	520.37 ± 0.25

TABLE II. Constants in viscosity-density power series fits ($\eta = a + bp + cp^2 + \dots$).

Gas	T (°C)	a (μP)	b ($\mu P\text{-cm}^3/g$)	c ($\mu P\text{-cm}^6/g^2$)	d ($\mu P\text{-cm}^9/g^3$)	e ($\mu P\text{-cm}^{12}/g^4$)	σ (μP)
N ₂	25	176.96 \pm 0.07	116.5 \pm 1.4	634 \pm 5			0.12
	-50	140.59 \pm 0.08	108.4 \pm 1.3	670 \pm 4			0.11
	-90	117.28 \pm 0.34	137.1 \pm 9.1	434 \pm 60	20 \pm 108		0.38
He	25	198.01 \pm 0.05	-41.1 \pm 9.8	2175 \pm 393			0.06
	-50	162.82 \pm 0.07	-35.2 \pm 10.0	1932 \pm 294			0.10
	-90	142.71 \pm 0.09	-46.2 \pm 11.0	2835 \pm 261			0.11
H ₂	25	88.55 \pm 0.03	144.8 \pm 12.3	7693 \pm 985			0.04
	-50	72.93 \pm 0.06	98.2 \pm 18.1	10531 \pm 1058			0.09
	-100	60.99 \pm 0.08	223.7 \pm 18.0	6148 \pm 843			0.10
Ar	25	225.30 \pm 0.12	109.0 \pm 3.8	442 \pm 31	-249 \pm 70		0.11
	-50	175.27 \pm 0.08	101.5 \pm 0.9	367 \pm 2			0.14
	-100	138.04 \pm 0.58	86.6 \pm 10.7	357 \pm 52	-82 \pm 93	212 \pm 54	0.61

TABLE III. Best estimates for first three virial coefficients of viscosity, from fits to subsets of the present data.

Gas	T (°C)	a (μP)	b (μP-cm ³ /g)	c (μP-cm ⁶ /g ²)
N ₂	25	176.96	116.5	633
	-50	140.49	111.6	655
	-90	117.52	122.7	603
Ar	25	225.17	116.2	359
	-50	175.19	103.2	363
	-100	137.83	91.0	338

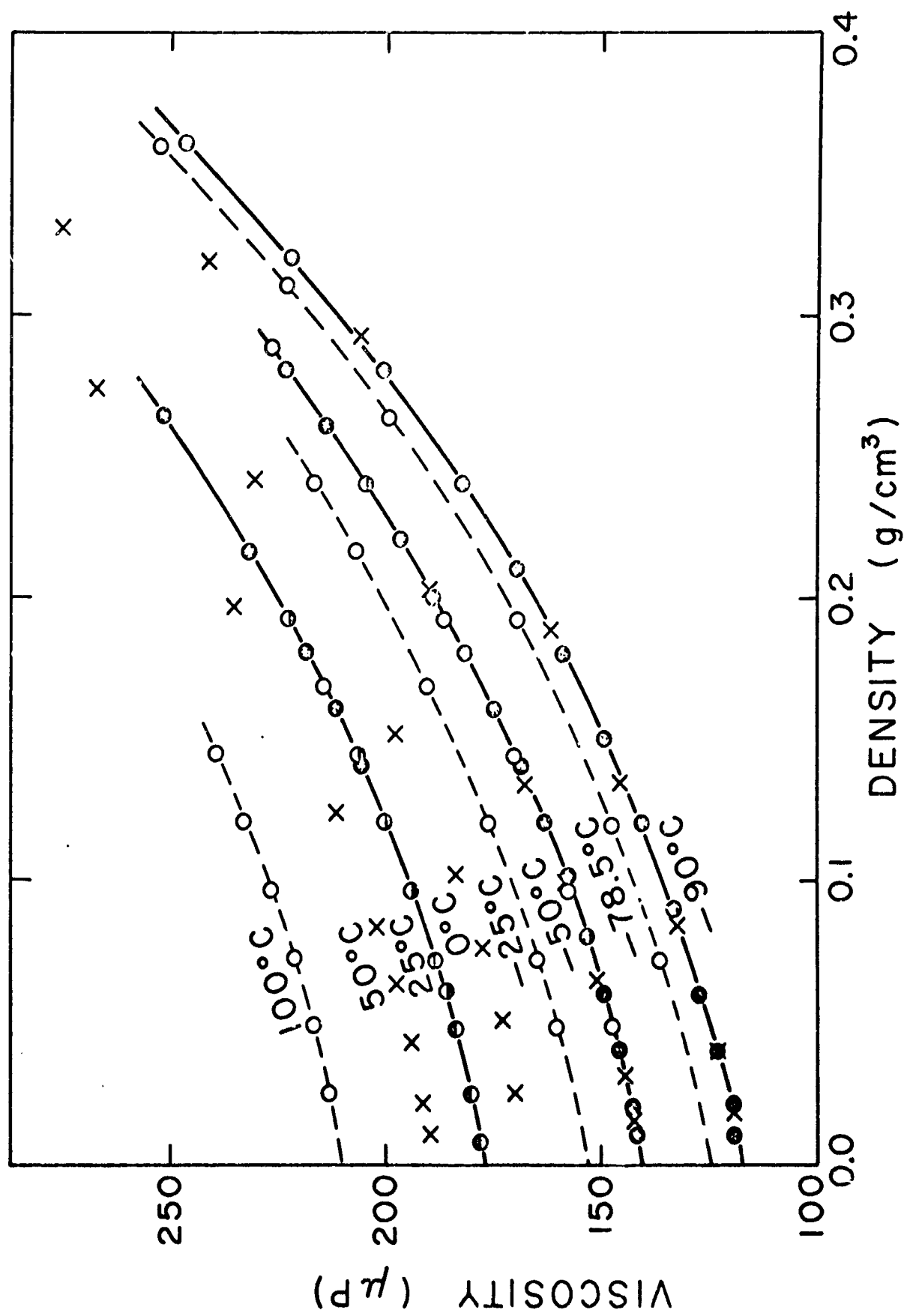
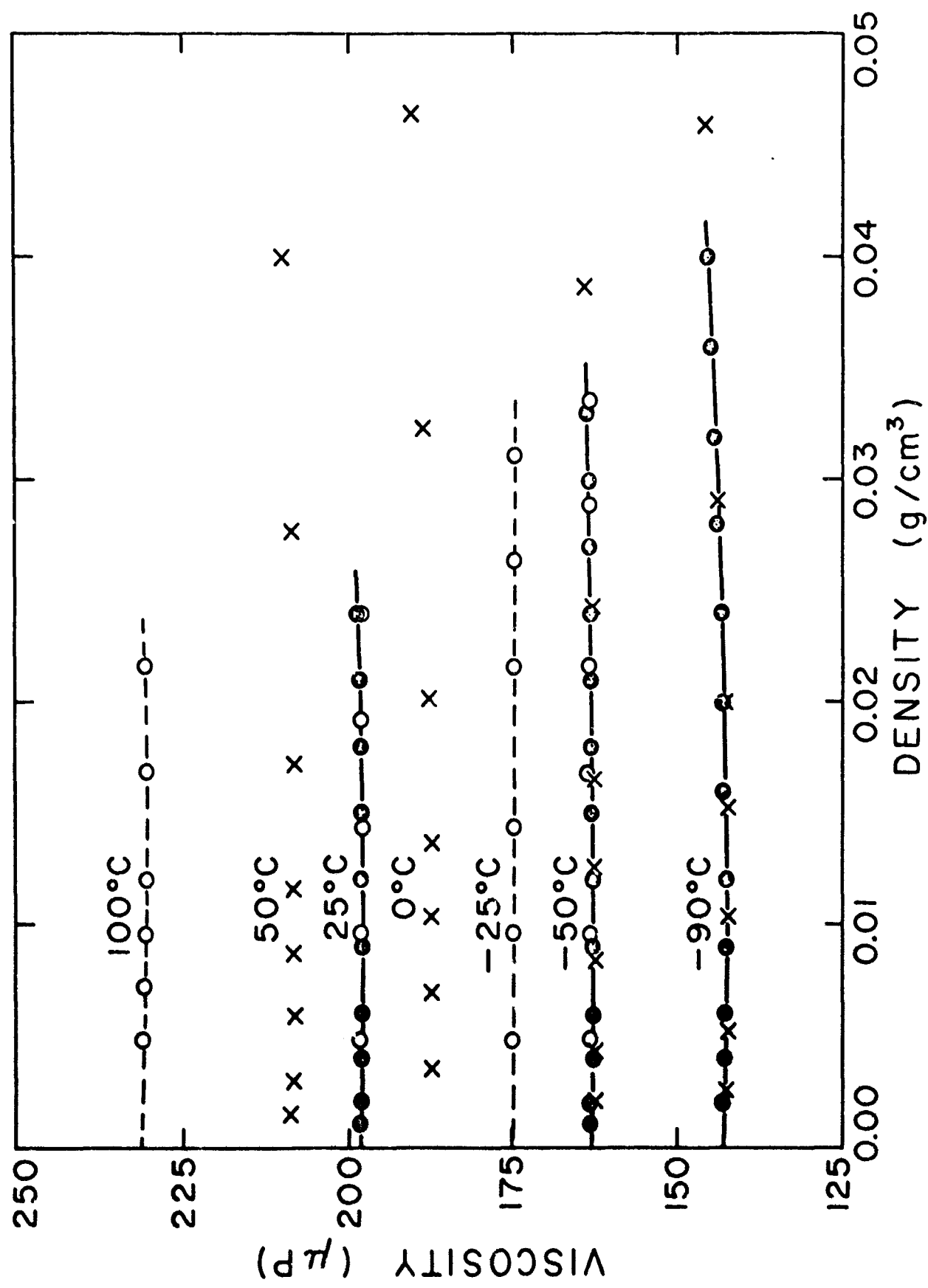


FIGURE 1.



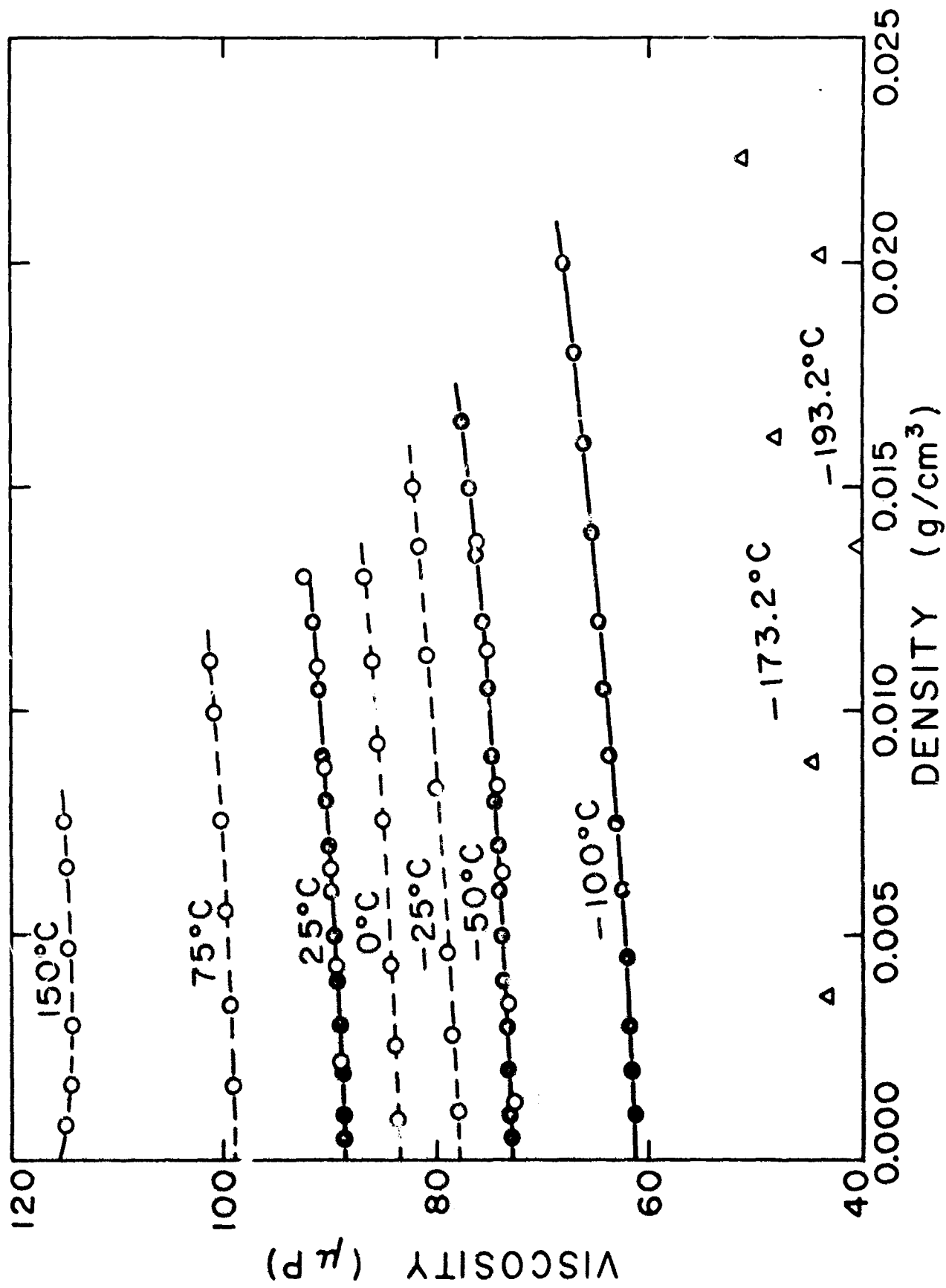


FIGURE 3.

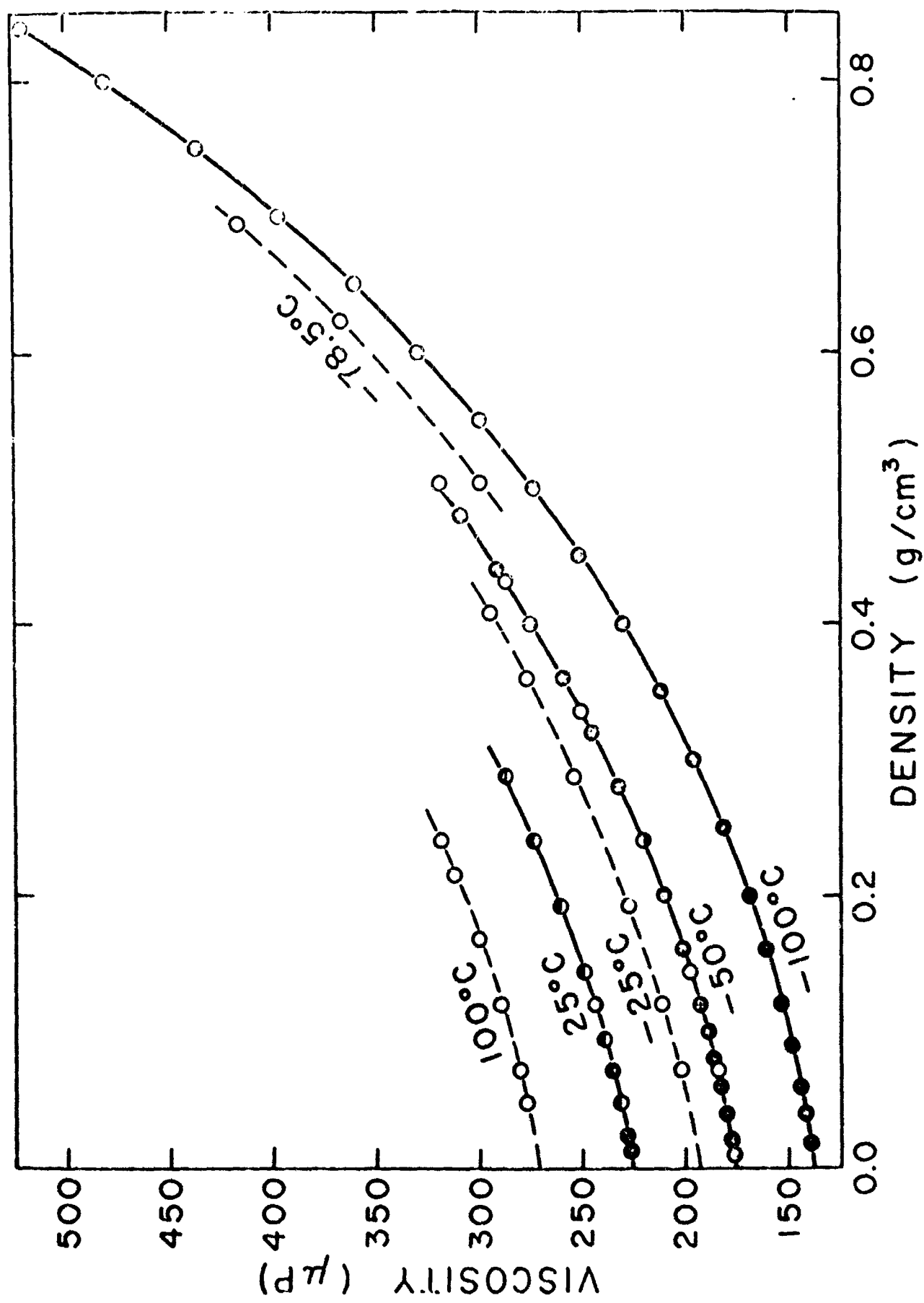


FIGURE 4.

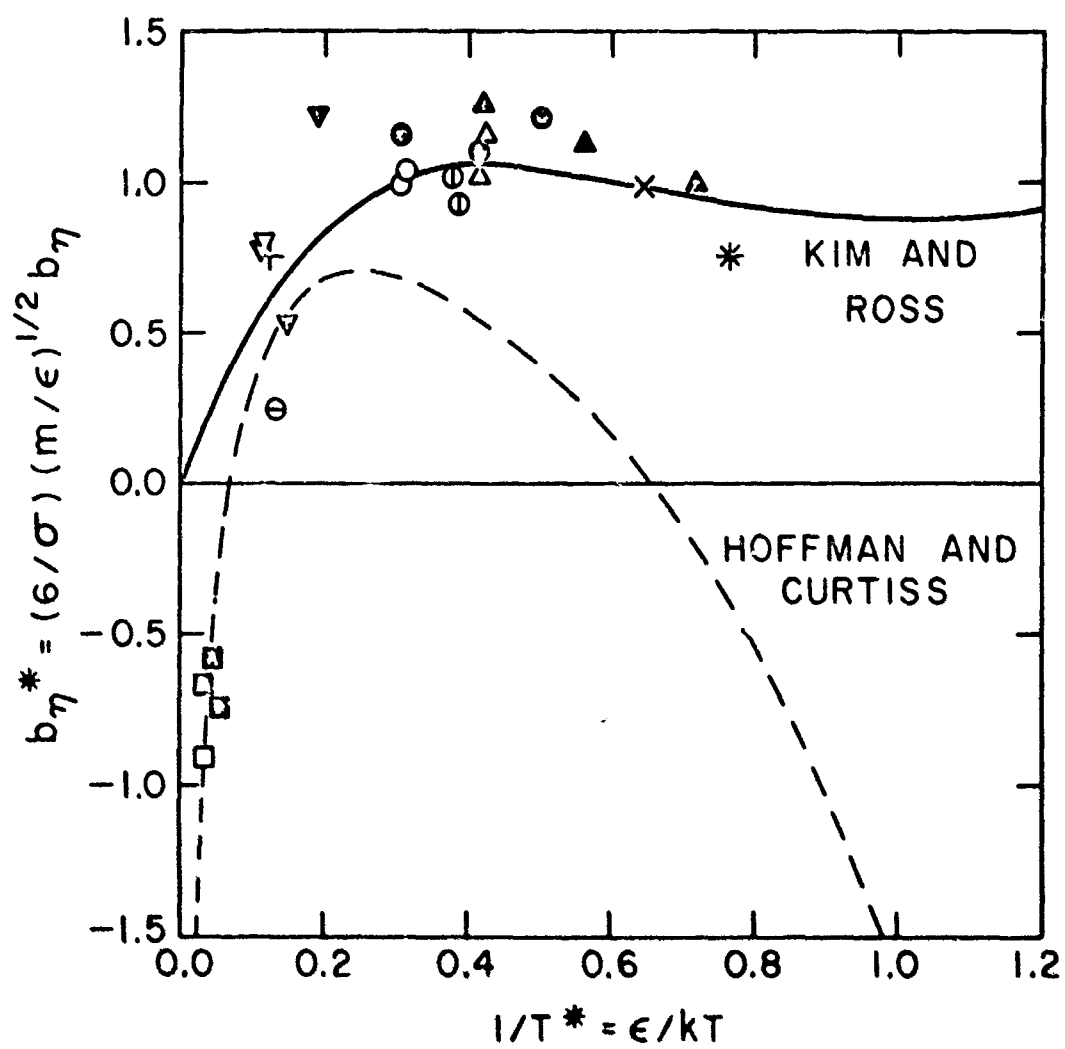


FIGURE 5.

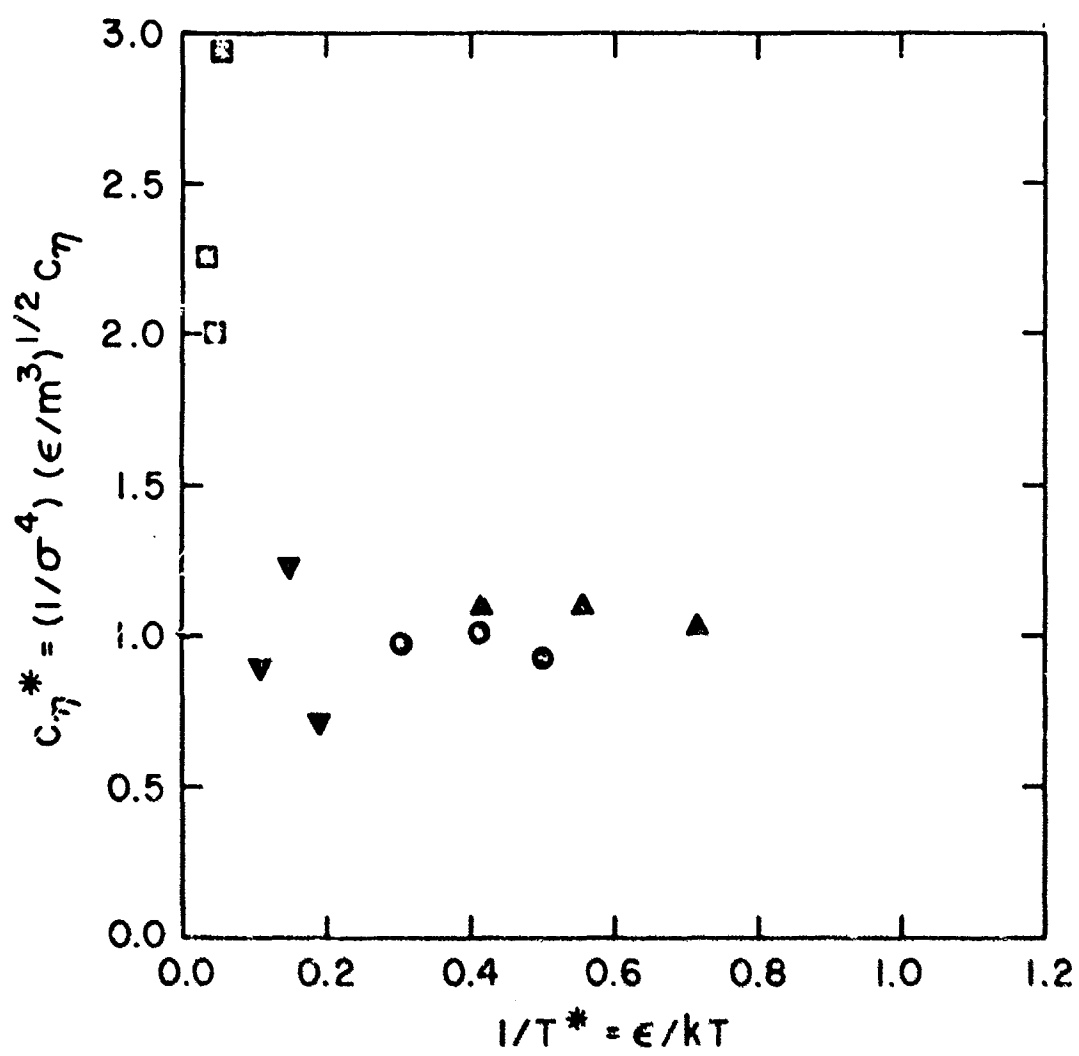


FIGURE 6.

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13. ABSTRACT			
<p>The range of a previously developed capillary-flow viscometer has been extended to -100°C, and the techniques of obtaining and determining the attainment of steady state have been improved. The viscosities of nitrogen, helium, hydrogen, and argon have been measured by an absolute method from -100° or -90° to 25°C and up to 150-175 atm (250 atm for N₂ at 25°C). The accuracy is estimated to be 0.1 - 0.2%, but is somewhat worse for Ar at -100°C. A number of empirical and theoretical analyses of the viscosity-density dependence have been made; while inconclusive, the evidence tends not to support the presence of a logarithmic term in the density expansion. Correlations of the second and the third virial coefficients of viscosity have been made.</p>			

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PAGE 11

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
<p>N₂, He, H₂, A</p> <p>viscosity-density dependence</p> <p>virial coefficients</p> <p>low temperature</p> <p>high pressure</p> <p>viscometer</p> <p>capillary flow</p>						

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